The effect of elastic inhomogeneity on equilibrium and stability of a two particle morphology

I. Schmidt a,*, R. Mueller b, D. Gross b

a Department of Engineering (Centre for Micromechanics), University of Cambridge, Trumpington Street, Cambridge CB2 1PZ, UK

b Institut für Mechanik, Technische Universität Darmstadt, D-64289 Darmstadt, Germany

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Abstract

The influence of elastic inhomogeneity on the precipitate morphology in a phase separating alloy, which is stressed due to the particle misfit, is investigated by calculating equilibrium shapes of two interacting inclusions in an orthotropic system. The method of computation is based on a variational formulation, involving Eshelby's notion of a 'force on an interface' and a boundary integral formulation of the associated linear elasticity problem. We find that the elastic inhomogeneity strongly affects both geometry and stability of equilibrium morphologies. Specifically, we find that, contrary to the homogeneous case, a two particle system can be an energetic minimum, implying the possibility of inverse coarsening. © 1998 Elsevier Science Ltd. All rights reserved.

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1. Introduction

The properties of phase separated alloys are, in general, closely connected to the microstructure of the material. Therefore, the microstructural evolution of such materials has been the subject of interest from both experimental and theoretical point of view. Phenomena that can be observed in these materials – a typical example being Nickel-base superalloys – include the evolution of γ' particles from spheres to cubes – eventually possessing concave interfaces – during growth, the alignment of these particles along crystallographic directions with increasing volume fraction, the formation of elongated precipitate shapes as well as the process of particle-splitting and the coalescence of precipitates, the latter resulting in raftlike microstructures (e.g. Maheshwari and Ardell, 1992, 1993; Meshkinpour and Ardell, 1994; Meshkinpour et al., 1994; Kaufmann et al., 1989; Pollock and Argon, 1994).

There is clear evidence that the above mentioned phenomena are closely related to the existence of an eigenstress in the system, owing to the elastically compensated misfit of the two distinct phases (e.g. Fährmann et al., 1995). This fact has been recognized in many theoretical investigations, a key part of which is the determination of the stress- and strainfields and the associated energy due to one or more misfitting inclusions with coherent interfaces in the framework of linear elasticity. In many works the knowledge of these fields is used to determine shapes or mutual arrangements of the particles that minimize – or reduce – the sum of elastic strain- and interfacial energy, in
order to characterize equilibrium states of the microstructure (for a detailed review see: Doi, 1996). In these calculations, the particle shapes are often constrained to be of a simple geometrical form, such as spheres, ellipsoids or cubes. However, they provided some qualitative explanations for e.g. the rafting behaviour of some alloys (Chang and Allen, 1991), the splitting of a cuboidal particle into two plates and further into an octet of cubes (Khachaturyan et al., 1988) or the occurrence of particle shape bifurcations (Johnson and Cahn, 1984; Johnson et al., 1988). The latter have been shown to exist also for the case where the particle shape is not constrained (Thompson et al., 1994), but the elastic moduli have been restricted to be identical in matrix and inclusion in this work. The influence of elastic inhomogeneity on this phenomenon has been studied by Schmidt and Gross (1995) and Schmidt and Gross (1997) and has been found to be of great importance.

Some authors focus on the temporal evolution towards equilibrium by prescribing an additional constitutive law, i.e. a rate equation for some microstructural parameter. Three different models have been utilized in these investigations, using different simplifying assumptions: first, a sharp interface model in which the microstructural evolution is described by an evolution law for the transient position of the phase separating interface (Voorhees et al., 1992; Su and Voorhees, 1996a, b), second, a continuously distributed concentration and order parameter as a field description of the microstructure, the evolution of which is governed by a Cahn-Hilliard- and Ginzburg-Landau-type equation respectively (Wang and Khachaturyan, 1995) and, third, a 'discrete atom model' in which the microstructure is represented through a large number of elementary points ('atoms') with pairwise interaction energy, using a Monte-Carlo-type simulation of the microstructural development (Lee, 1996a, b). Su and Voorhees (1996b) found that a long-range attractive- and a short-range repulsive force acts between two misfitting particles, the latter preventing coalescence of the particles. Further, they found that inverse coarsening appeared only at intermediate stages of the process and that a two- or three particle system is never stable with respect to coarsening, i.e. cannot represent an equilibrium configuration.

Focusing on the growth process of a single particle, the calculations of Wang and Khachaturyan (1995) suggest that the occurrence of concave interfaces is most likely a kinetic effect that vanishes in the equilibrium state. On the other hand, particles with concave interfaces have been found by Schmidt and Gross (1997) to be accessible equilibrium morphologies when the elastic inhomogeneity is taken into account.

While the above mentioned results are limited to the case of homogeneous elastic moduli in matrix and precipitate, this restriction is dropped in the calculations of Lee (1996a, b). However, due to the specific modelling of anisotropic elasticity, the elastic constants of both matrix and inclusion are confined to obey Cauchy's rule, i.e. $C_{12} = C_{44}$.

Some aspects of the influence of elastic inhomogeneity on particle interaction have been addressed in this work, such as the coalescence of soft- and the stability of hard particles. Apart from the restriction placed on the elastic moduli, results have been obtained only for the case where the elastic constants of the two phases are a multiple of each other – i.e. $C_{ij}$ (matrix) = $\alpha C_{ij}$ (inclusion) – thus the anisotropy is the same in both phases and, furthermore, is kept constant in all calculations.

The results of Lee (1996a, b) as well as other investigations, which used a fixed particle morphology, revealed a strong influence of the elastic inhomogeneity on the kinetics and equilibrium of multiparticle systems. However, no detailed study on the influence of all elastic constants and the interfacial energy on the particle interaction seems to have been published, to the best of the authors knowledge. It is this question that shall be addressed in this paper. To this end, we calculate energy extremizing morphologies of an elastically inhomogeneous two particle system – and their stability – under plain strain conditions, using a sharp interface model and taking into account an isotropic, deformation-independent interfacial energy density.

The strategy for determining equilibrium configurations has been presented in some detail by Schmidt and Gross (1997). Therefore, only the essentials of the method are briefly recalled here,
as well as some results for the case of a single inclusion, because they bear some relevance to the results to be presented here.

2. Problem formulation

2.1. Basic equations

We consider an infinitely extended matrix, \( \mathcal{K}^+ \), containing a number of inclusions, \( \mathcal{K}_m^-; m = 1 \ldots K \), of a given total area, whose phase interfaces are endowed with an isotropic, deformation-independent interfacial energy density \( \gamma \). Fig. 1 shows the situation for a two particle arrangement. Both phases are assumed to be linearly elastic, having distinct elastic constants

\[
C(x) = \begin{cases} 
C^+ & \text{in } \mathcal{K}^+ \\
C^- & \text{in } \mathcal{K}_m^- \end{cases}, \quad m = 1 \ldots K \tag{1}
\]

Under plain strain conditions and when the phases are taken to be homogeneous, the systems free energy is the sum of the elastic strain energy and the interfacial energy and is given by

\[
\Psi[u, \partial\mathcal{K}_1^-, \ldots, \partial\mathcal{K}_K^-] = \frac{1}{2} \int_{\mathcal{K}} \sigma : \varepsilon \, dA + \sum_{m=1}^{K} \oint\gamma \, d\ell, \tag{2}
\]

where \( \mathcal{K} = \mathcal{K}^+ \cup \bigcup_{m=1}^{K} \mathcal{K}_m^- \) is the area occupied by the whole system,

\[
\sigma = C(x)\varepsilon \tag{3}
\]

is the stress tensor and \( u \) denotes the displacement field. Taking the stress free state of the matrix phase as a reference state, the displacement field is continuous,

\[
[u] = 0 \text{ on } \partial\mathcal{K}_m^-, \quad m = 1 \ldots K \tag{4}
\]

and the elastic strain is given by

\[
\varepsilon_{el}(x) = \begin{cases} 
\varepsilon(x) & \text{in } \mathcal{K}^+ \\
\varepsilon(x) - \varepsilon^0 & \text{in } \mathcal{K}_m^-, \quad m = 1 \ldots K \end{cases} \tag{5}
\]

where \( \varepsilon = \frac{1}{2}(\nabla u + \nabla u^T) \) is the total strain tensor and \( \varepsilon^0 \) is the transformation strain tensor – the latter being the same in all inclusions.

On thermodynamic grounds, the equilibrium shape will be such that it minimizes \( \Psi \) under the constraint that the total area of the inclusions be equal to a given quantity \( A_0 \):

\[
\sum_{m=1}^{K} A_m = A_0. \tag{6}
\]

Requiring \( \Psi \) to be stationary with respect to the displacement field gives the mechanical equilibrium conditions

\[
\text{div } \sigma = 0 \text{ in } \mathcal{K}, \tag{7}
\]

\[
[[\sigma]]n = 0 \text{ on } \partial\mathcal{K}_m^-, \quad m = 1 \ldots K, \tag{7}
\]

in which \( n \) is the unit outward normal on the respective interface \( \partial\mathcal{K}_m^- \) and \( [[ \cdot ]] = (\cdot)^+ - (\cdot)^- \) denotes the jump of a quantity across the interface. Stationarity with respect to the interface positions demands

\[
\tau_n n = 0 \text{ on } \partial\mathcal{K}_m^-, \quad m = 1 \ldots K, \tag{8}
\]

where (cf. Eshelby, 1970; Leo and Sekerka, 1989; Schmidt and Gross, 1997)

\[
\tau_n = \frac{1}{2} (\varepsilon + \varepsilon^0) \cdot \Gamma(n)(\varepsilon + \varepsilon^0) + \frac{1}{2} \varepsilon^0 \cdot A \varepsilon^0 - \gamma \kappa + \lambda, \tag{9}
\]

is the driving force acting on the interface. The following abbreviations have been used:

\[
\Gamma(n) = [[C]] - [[C]](n \otimes \Omega^{-1}(n) \otimes n)[[C]],
\]

\[
A = -([[C]] + [[C]](C^-)^{-1}[[C]]) = \text{const.},
\]

\[
\Omega_{ik} = C_{ik,j}n_j n_i, \quad i,j = 1, \ldots, D, \tag{10}
\]

\[
\varepsilon^0 = [[C]]^{-1} C^- \varepsilon^0.
\]
In Eq. (9) \( \kappa \) is the interfacial curvature \(^1\), \( \varepsilon \) is the limiting value of the total strain when approaching the interface from inside the inclusion (i.e. the minus-side) and \( \lambda \) is the Lagrange-multiplier that results from the incorporation of the side condition (6) into the variational formulation. The latter can be interpreted as a chemical driving force, because its role is identical to that of a deformation independent part of the phases' free energy (cf. Schmidt and Gross, 1997). Eqs. (7) and (8) are the necessary conditions for the equilibrium morphology we are looking for. The method for determining the stability of a solution is described later in the Section on numerics.

2.2. Non-dimensionalization

In order to present results sensibly, the following non-dimensionalization is used: The characteristic length \( \ell^* = \sqrt{\sum_{m=1}^{K} A_m} \) is used to scale the position- and displacement vectors \( x \) and \( u \), as well as the particle areas \( A_m \) and the interfacial curvature \( \kappa \). A characteristic stiffness is given by the average shear modulus of the matrix material, \( \mu^* = \frac{1}{2\pi} \int_0^{2\pi} C_{1212}(\varphi) d\varphi \), which is used to scale the elastic constants tensors \( C^e \) as well as the tensors \( \Gamma \) and \( A \) in Eq. (10). For the isotropic eigenstrains to be considered in the two-particle case, \( \varepsilon^0 = \varepsilon^* \mathbf{1} \), the quantity \( \varepsilon^* \) serves to scale the strains \( \varepsilon, \varepsilon_{\text{ed}} \) and to define a typical energy of the system through \( \Psi^* = \gamma \ell^* + \mu^* \varepsilon^*(\ell^*)^2 \). With this choice, the total energy of the system and the driving force are expressed, respectively, as

\[
\Psi = \frac{1}{2} \frac{L}{1 + L} \int_{\partial I} \sigma \cdot \varepsilon_{\text{ed}} \, dA + \frac{1}{1 + L} \sum_{m=1}^{K} \int_{\partial I_m} \varepsilon \, d\ell,
\]

\[
\tau_n = \frac{L}{1 + L} \left\{ \frac{1}{2} (\varepsilon + \varepsilon^0) \cdot \Gamma(n)(\varepsilon + \varepsilon^0) + \frac{1}{2} \varepsilon^0 \cdot \Lambda \varepsilon^0 + \lambda \right\} - \frac{\kappa}{1 + L},
\]

where all quantities are dimensionless and

\[
L = (\ell^* \mu^* (\varepsilon^*)^2)/\gamma \tag{12}
\]

can be understood as the dimensionless size of the particle ensemble. The magnitude of \( L \) reflects the relative importance of the elastic energy as compared to the interfacial energy.

3. Numerical method

3.1. Boundary-integral technique

To calculate the generalized driving force \( \tau_n \) on the interfaces the elastic field equations have to be solved. Analytical solutions are only available for a limited number of geometries and material properties. The classical solutions by Eshelby, see for example Mura (1987), are restricted to ellipsoids/ellipses and isotropic materials. In order to treat orthotropic or anisotropic material together with arbitrary geometries, a Boundary Element Method (BEM) is used. The BEM can be viewed as a natural choice of a numerical method, as it allows the use of equations on the interfaces only, thus reducing the problem-dimension by one.

While we restrict our attention to the 2D plain strain problem here, the method can also be applied in the 3D case (see Mueller and Gross (1997) for a discussion of the equilibrium morphology of a single particle).

Applying the standard Boundary Integral Equation (BIE) to the multiple inclusion problem, we get a set of integral equations. For the \( K \) inclusions the \( K \) integral equations read as follows

\[
e(x)u_m(x) + \int_{\partial \omega_m} T^- (x,y) [u_m(y) - u_{m0}^0(y)] d\ell
\]

\[
= \int_{\partial \omega_m} U^- (x,y) t_m(y) d\ell \quad \text{for} \quad m = 1, \ldots, K, \tag{13}
\]

where \( u_m \) and \( t_m \) stand for the displacements and tractions on the \( m \)th interface, while \( U^- \) and \( T^- \) denote the corresponding fundamental solutions of the inclusion material. The displacements due to the inelastic eigenstrains \( \varepsilon^0 \) are denoted by \( u_{m0}^0 \). For the plane strain state, the fundamental solutions \( U \) and \( T \) of orthotropic or general anisotropic materials

\(^1\) i.e. twice the mean curvature, taken to be positive for a sphere.
are available in closed form (Green, 1943). With the fundamental solutions of the matrix material, denoted by $^+$, the BIE of the matrix is given by

$$c(x|u(x) + \int_{\partial\mathcal{K}^+} T^+(x,y)u(y)\mathrm{d}\ell$$

$$= \int_{\partial\mathcal{K}^+} U^+(x,y)t(y)\mathrm{d}\ell \quad \text{where} \quad \partial\mathcal{K}^+$$

$$= \bigcup_{m=1}^K \partial\mathcal{K}^+, \quad (14)$$

Note that in contrast to Eq. (13), in which the integration is performed on each interface separately, in Eq. (14) the integrals are evaluated on the complete interface area. This is indicated by dropping the index $m$ on $u$ and in Eq. (14) the area of integration by $\partial\mathcal{K}^+$.

The above BIE’s (13) and (14) are discretized with $N$ two-node boundary elements on each interface. The nodal displacements $u_p$ and nodal tractions $t_p$ on each interface are interpolated with piecewise linear shape functions of the contour arc length $\Phi_p(x)$

$$u \approx \sum_{p=1}^N \Phi_p(s)u_p, \quad t \approx \sum_{p=1}^N \Phi_p(s)t_p. \quad (15)$$

Together with the continuity conditions (4) and (7), insertion of this interpolation in the two BIE’s (13) and (14) leads to a linear system of equations,

$$\begin{bmatrix}
A^+ & B^+ \\
A_1^- & 0 & 0 & -B_1^- & 0 & 0 \\
0 & \vdots & 0 & 0 & \vdots & 0 \\
0 & 0 & A_K^- & 0 & 0 & -B_K^- \\
\end{bmatrix}
\begin{bmatrix}
u_1^h \\
\vdots \\
u_K^h \\
t_1^h \\
\vdots \\
t_K^h \\
\end{bmatrix} = 
\begin{bmatrix}
0 \\
\vdots \\
0 \\
A_1^-u_1^o \\
\vdots \\
A_K^-u_K^o \\
\end{bmatrix}, \quad (16)$$

where $u_m^o$, $t_m^o$ stands for the vector of the nodal displacements and -tractions on the $m$th interface respectively.

The linear system (16) reflects the fact that the multiple inclusion problem is coupled only through the matrix material. The right hand side of the system (16), i.e. the loading, is given by the inelastic displacements $u^o$ arising from the eigenstrain $\varepsilon^0$. The solution of (16) gives the nodal displacements $u_m^o$ and nodal tractions $t_m^o$ on all interfaces $\partial\mathcal{K}^m (m = 1, \ldots, K)$. The strains $\varepsilon$ and the generalized driving force $\tau_n$ are then calculated in a straightforward manner (see Schmidt and Gross (1997) for further details).

### 3.2. Equilibrium morphology

With the boundary-element discretization of the interface, the question now becomes to determine the position of the nodal points such that Eq. (8) is satisfied. That is, the discretization used for the boundary-element method is at the same time the approximation for the solution of Eq. (8).

With $x_p$ being the distance of the nodal point number $p$ from a fixed point $x_i$ in the fixed direction $h_p, ||h_p|| = 1$, the approximation of each of the interfaces may be written as

$$x^{(m)}_n(s) = x_n^{(n)} + \sum_{p=1}^N z_p^{(m)} h_p(m) \Phi_p^{(m)}(s),$$

$$m = 1 \ldots K. \quad (17)$$

Eq. (8) is now treated with a Galerkin method. The residue of the left-hand side of Eq. (8), obtained upon inserting Eq. (17) into Eq. (8), is weighted with the ansatz-functions and these weighted residues are then forced to vanish. With $z_p^{(m)}$, $h_p^{(m)} \Phi_p^{(m)}$ being the free parameters and the ansatz-functions respectively, the Galerkin method yields

$$\int_{\partial\mathcal{K}^{m}} z_p^{(m)}(\alpha^{(m)}_1, \ldots, \alpha^{(m)}_N, \lambda) = \int_{\partial\mathcal{K}^{m}} \tau_n u \cdot h_p^{(m)} \Phi_p^{(m)} \mathrm{d}\ell = 0,$$

$$p = 1 \ldots N, \quad m = 1 \ldots K. \quad (18)$$

Together with the side condition (6), expressed in terms of boundary integrals,
\[ g_{KN+1}(x_1^{(1)}, \ldots, x_N^{(1)}, \ldots, x_1^{(K)}, \ldots, x_N^{(K)}, \lambda) = \frac{1}{2} \sum_{m=1}^{K} \int_{\Omega_{Q_m}} x_n \, d\ell - A_0 = 0 \]  

This represents a set of \( KN + 1 \) non-linear equations for the \( KN \) unknown shape parameters \( x_p^{(m)} \) and \( \lambda \). Since we are dealing with a conservative system, the above described procedure can also be understood as a Ritz method, where the free energy is extremized with respect to the \( x_p^{(m)} \).

### 3.3. Stability

The stability of a solution \( x^*, \lambda^* \) is examined by an eigenvalue analysis of the Hessian

\[ H_{pq} = -\frac{\partial^2 g_p}{\partial x_q^2} \bigg|_{x^*, \lambda^*}, \quad p, q = 1, \ldots, KN \]  

which is computed by numerical differentiation. The sign of the smallest eigenvalue, whose corresponding eigenvector does not violate the side condition (6), indicates the stability of a solution: a positive value implies a minimum of the energy and therefore a stable solution.

### 4. Results

#### 4.1. Material parameters

The results to be presented in the sequel apply for a system of two distinct orthotropic materials, each having cubic symmetry. These materials are characterized by (i) an average stiffness \( \bar{\mu} \), (ii) an anisotropy ratio \( A \) and (iii) an average Poisson number \( \bar{\nu} \). The relation of these numbers with the classical Voigt constants \( C_{ij} \) is given in the Appendix. With this choice, the equilibrium morphology depends on six dimensionless quantities, i.e.

\[ L, \bar{\mu}^-/\bar{\mu}^+, A^+, A^-, \bar{\nu}^+, \bar{\nu}^- \]  

The influence of these parameters is examined by computing equilibrium morphologies for \( L \) continuously varying between 3...20, while the other parameters are discretely varied, one at a time, around a ‘ground state’. This state is the homogeneous case, given by

\[ \bar{\mu}^-/\bar{\mu}^+ = 1.0, \quad A^+ = A^- = 2.5, \quad \bar{\nu}^+ = \bar{\nu}^- = 0.3 \]

and is meant to mimic the situation in Nickel-base superalloys.

#### 4.2. Single particle results

For a better understanding of the results to follow, we briefly recall some basic results for the case of a single inclusion (Thompson et al., 1994; Schmidt and Gross, 1997). For small \( L \), the interfacial energy prevails and the equilibrium shape is circle-like. With increasing \( L \), the elastic energy becomes more important and the shapes become more square-like, their square sides being oriented along the [0 0 1] and [0 1 0] direction. At a critical value of \( L \), \( L_{\text{crit}} \), these fourfold symmetric shapes become unstable: the stable equilibrium shapes in that regime assume a twofold symmetry and are elongated in the [0 0 1] or [0 1 0] direction. The situation is illustrated in Fig. 2 which shows the typical diagram of a symmetry breaking bifurcation in terms of the geometric quantity

\[ \frac{\bar{\mu}^-}{\bar{\mu}^+} = 1.0, \quad A^+ = A^- = 2.5, \quad \bar{\nu}^+ = \bar{\nu}^- = 0.3 \]

and is meant to mimic the situation in Nickel-base superalloys.

![Fig. 2. Bifurcation diagram for the single particle case: Aspect ratio vs. particle size.](image)

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\[^2\] Since the equilibrium shape depends on both, stresses and strains in the bimaterial, there is no reduced dependence on some anisotropic generalization of Dundurs constants.
\( \bar{\rho} = (\rho - 1)/(\rho + 1) \), where \( \rho \) is the aspect ratio of the shape. This shape instability is caused by a redistribution of the energies in matrix, inclusion and interface: the increase of interfacial energy due to the enlarged interface of the elongated shape and the increase of the elastic strain energy in the inclusion is compensated by the saving of elastic strain energy in the matrix.

The critical value \( L_{\text{crit}} \) has been found to depend sensitively on the material parameters of both matrix and inclusion. This manifests itself in the steep gradients of some of the curves in Fig. 3 which show \( L_{\text{crit}} \) as a function of the parameters (21).

If the symmetry of the system is disturbed by an external uniaxial load or a non-isotropic eigenstrain \(^3\), the bifurcation diagram assumes the form shown in Fig. 4. \(^4\) One family of solutions exists throughout the whole range of \( L \)-values. The respective particle shapes are elongated in the direction of the smaller of the two principal values of the eigenstrain; these shapes represent stable solutions. Two other families of solutions exist above a certain value of \( L \), both being elongated in the

\[ \bar{\mu}^-/\bar{\mu}^+ = 1.0, \quad \epsilon_1^0/\epsilon_2^0 = 0.95 \]

\[ \bar{\rho} \rightarrow 0 \]

\[ L \]

\[ \bar{\mu}^+/\bar{\mu}^- \]

\[ A^- \]

\[ A^+ \]

\[ \bar{\nu}^- \]

Fig. 3. Critical particle size for shape instability vs. material parameters.

Fig. 4. Disturbed bifurcation diagram for tetragonal eigenstrain.

\(^3\) These two cases are equivalent and can be replaced by one another, cf. Schmidt (1997).

\(^4\) In this case, the larger of the two principal eigenstrains is used as \( \epsilon_1^0 \) for the non-dimensionalization described in Section 2.2.
perpendicular direction. The upper branch represents unstable - the lower branch stable solutions.

4.3. Two interacting particles

4.3.1. The influence of the stiffness ratio

In the case of a two (or multi-) particle system, each particle experiences an – non-uniform – ‘external stressfield’ which is induced by the misfit of the other particles. Therefore the results can be expected to be qualitatively similar to the diagram Fig. 4. Indeed, we find the three different families of shapes described in the last Section, that satisfy Eq. (8). The aspect ratio of these shapes is plotted against \( L \) in Fig. 5 for the homogeneous case, \( \bar{\mu}^-/\bar{\mu}^+ = 1 \), together with the corresponding shapes. With increasing \( L \), the shapes of the first family become more elongated and move closer together. Above \( L \approx 18 \) there exist two more solution families. The shapes of both of these families are elongated in the [0 1 0] direction, their elongated sides facing each other. The particles of the lowermost branch are considerably farther apart than those of all the other families.

The eigenvalue analysis shows that all three solution types are unstable in the considered \( L \)-interval. This is different for the case of a stiffer inclusion phase, \( \bar{\mu}^-/\bar{\mu}^+ = 1.2 \). Here, the stability-indicating eigenvalue for the shapes of the first solution family passes through zero at \( L \approx 16 \), this family being the only existent in the considered interval of \( L \) (Fig. 6). Fig. 7 depicts the four smallest eigenvalues as a function of \( L \) together with the corresponding eigenforms (i.e. the eigenvector of the Hessian added to the respective solution \( x^* \)). The eigenform of the lowermost curve obviously violates the side condition of fixed total particle area. The corresponding eigenvalue is therefore irrelevant. The second-smallest eigenvalue is the one that indicates the stability of the solution, the corresponding eigenform being es-
sentially the growth of one particle at the expense of the other. This eigenvalue passes through zero for $L \approx 12$, which is not immediately visible as the curves of the second and third eigenvalue seem to veer each other there. This ‘curve-veering’ phenomenon is related to the numerical calculation of the Hessian and its eigenvalues and has been observed also in connection with the calculation of eigenfrequencies of elastic plates (Leissa, 1974; Kuttler and Sigillitto, 1981). In our case, the third eigenvalue is a zero-eigenvalue independently of $L$, as the corresponding eigenform is a simple rigid body motion of the two particles as a whole. Because the eigenforms of the second and third smallest eigenvalues have interchanged for $L > 12$ (not shown in the figure), this means that one eigenvalue must have passed through zero and, thus, the corresponding solution is stable.

This result is confirmed by directly calculating the energy of equilibrium configurations with two particles of unequal size, where the size of each individual particle is prescribed but the shapes are otherwise unconstrained. In Fig. 8 the energy of these configurations is plotted as a function of $(1 - \alpha)/(1 + \alpha)$ for different $L$ ($L = 9/12/15$), where $\alpha = A_1/A_2$ is the ratio of particle areas. It can be seen that for $L = 15$ the equally sized particle configuration is a minimizer, while for $L = 9$ it is a maximizer. In the transition regime for $L = 12$, the energy exhibits an extended plateau. Altering the scaling such that these three curves can be compared in one plot, makes clear that, regarding the energy, there is hardly any difference between all the configurations for fixed $L$ (Fig. 8, top left).

While for $\bar{\mu}^-/\bar{\mu}^+ = 1.2$ the solutions of the second and third type do not exist in the considered $L$-interval, for $\bar{\mu}^-/\bar{\mu}^+ = 0.8$ (i.e. the matrix is stiffer than the inclusions) these solutions occur at smaller $L$, while the solutions of the first type do not exist for any value of $L$ (Fig. 9). This is because the particles have the tendency to merge. Fig. 10 illustrates how the decrease of the stiffness ratio reduces the shortest distance between the interfaces of the two particles. For $\bar{\mu}^-/\bar{\mu}^+ = 0.8$ there is no repulsive force between the particles, irrespective of how close their interfaces are.

Here, the eigenvalue analysis reveals that all the solutions shown in Fig. 9 are unstable. The
The smallest eigenvalues together with the corresponding eigenforms are shown in Fig. 11 for the second solution family (the upper branch in Fig. 9). Like in the single particle case, these shapes are unstable with respect to changes of their aspect ratio. Fig. 11 makes also clear that these solutions cannot be expected to become stable for some larger value of $L$.

### 4.3.2. The influence of the matrix’ anisotropy

The effect of altering the anisotropy of the matrix material, $A^+$, is shown in Fig. 12. Like before for $\bar{\mu}/\bar{\mu}^+ = 0.8$, the first solution family does not exist for $A^+ = 1.5$ and it ceases to exist at $L \approx 6$ for $A^+ = 2.0$. Just as a decrease of the stiffness ratio, a decrease of $A^+$ reduces the shortest distance between the particles. Fig. 13 illustrates, how the distance tends to zero for $A^+ = 2.0$. As could be expected from the influence of $A^+$ on $L_{\text{crit}}$, a decrease of the matrix’ anisotropy makes the existence of the second and third solution type possible at smaller $L$-values (Fig. 12).

Fig. 14 depicts the smallest eigenvalues together with the corresponding eigenforms as a function of $L$ for the third solution family (the lower branch in Fig. 10. Shortest interface distance vs. particle size for different stiffness ratios. Fig. 9. Aspect ratio vs. particle size: Matrix stiffer than inclusion.

Fig. 11. Eigenvalues and corresponding eigenforms for the second-type solutions for $A^+ = 1.5$.

Fig. 12. Aspect ratio vs. particle size for different matrix-anisotropies.

Fig. 13. Shortest interface distance vs. particle size for different stiffness ratios.
4.3.3. The influence of the inclusions anisotropy

The influence of the anisotropy of the inclusion material, $A^-$, is shown in Fig. 15. Regarding the aspect ratio, its effect is just opposite to that of $A^+$ – this being in agreement from what could be expected from the single particle results (Fig. 3). For $A^-=3$ the particles of the first solution family tend to coalesce for $L > 6$ and solutions of the second and third kind exist for $L > 12.5$. Decreasing $A^-$ leads to smaller aspect ratios for the shapes of the first type solutions and can even produce shapes that are elongated in [0 1 0] direction, i.e. $\hat{\bar{q}} < 0$. These are shown in Fig. 16. For $A^- = 1.5$, the eigenvalue analysis yields that the solutions are stable for $L > 14$, because the relevant eigenvalue passes trough zero there (Fig. 17, solid line). Fig. 18 depicts the smallest interface distance as a function of $L$, the arrow indicating

Fig. 13. Shortest interface distance vs. particle size for different matrix-anisotropies.

Fig. 14. Eigenvalues and corresponding eigenforms for the third-type solutions from Fig. 9.

Fig. 15. Aspect ratio vs. particle size for different inclusion-anisotropies.

Fig. 16. Aspect ratio vs. particle size: First-type solutions with $\hat{\bar{q}} < 0$.
increasing values of $A^-$. It can be seen, how the distance tends to zero for $A^- = 3$ and that the interparticle distance is increasing with $L$ for $A^- = 1.5$ – this being in contrast to all other results found here.

4.3.4. The influence of the inclusions Poisson number

For completeness, Figs. 19 and 20 show the results for different values of the average Poisson number $\bar{\nu}^- (\bar{\nu}^- = 0.2/0.3/0.4)$. Considering that, for most materials of cubic symmetry, $\bar{\nu}^-$ is fairly close to 0.3, its influence is less pronounced as compared to the other material parameters. Note that no particle coalescence occurs here for all values of $\bar{\nu}^-$. While increasing $\bar{\nu}^-$ produces larger values of the smallest eigenvalue, the latter remains negative for all the curves shown in Fig. 19.

Like in the case of a single inclusion, it was found here, that the average Poisson number of the inclusion material, $\bar{\nu}^+$, has practically no influence on the equilibrium morphology.

5. Discussion

The three different kinds of equilibrium morphologies that have been found for two interacting
particles can be understood by considering the bifurcation diagram for an isolated particle (Fig. 4) and viewing the second particle as an imperfection of the otherwise fourfold symmetry. The equilibrium morphology can therefore have, at most, two axes of symmetry – and at least one solution will, as is known from bifurcation theory (Troger and Steindl, 1991). While all the equilibrium configurations presented in the previous Section are comprised of two mirror-symmetry-related particles, they represent local extrema of the functional (2) with no constraint placed on their shapes (other than the given total area). The stability analysis also does not require any symmetry conditions of the solutions.

The first type solutions exist for small \( L \) (i.e. small particles or large interfacial energy density). The shape of either one of the two particles is essentially that which is produced by an eigenstrain with a small deviatoric part, i.e. \( \varepsilon^1_1/\varepsilon^0_2 = 0.95 \) (compare Fig. 4 and, e.g., Fig. 5, where the \( L \) value from the latter has to be divided by \( \sqrt{2} \), because the size of one of the two particles is only half of that of the single one).

Just like in the single particle case, the shapes become more elongated with increasing \( L \), because the elastic energy, that favors a plate-like shape, becomes increasingly important relative to the interfacial energy. Two additional solution families exist above a certain value of \( L \). These correspond to the isolated curve in the disturbed bifurcation diagram of the single particle (Fig. 4). Generally, the particles move closer together with increasing \( L \), although there are exceptions from this rule (see Fig. 18). This is essentially due to the fact that, for small \( L \), a larger share of the interface deviates from the [0 0 1] or [0 1 0] direction (which would be preferred from the elastic energy) in order to minimize the interface area. The strains produced by these more sphere-like shapes are such that it becomes disadvantageous to put them close together. One might say that only if the interfaces are allowed to align and thereby straighten up, i.e. for larger \( L \), does it become energetically favorable to move them closer together.

For the range of parameters considered here, most of the equilibrium morphologies we found are unstable. Those of the first and third solution family are unstable with respect to a change in the particle sizes, the ones of the second family are unstable with respect to a change in their aspect ratio. However, there is a regime of realistic parameter-values for which the first type solutions become stable above a certain particle size \( L \). This value has to be sufficiently large, as the interfacial energy always favors one single particle over two separated ones, this being known from the classical Ostwald-ripening. There has been some discussion about whether or not a two particle system can be an energetic minimum, implying that inverse coarsening, i.e. the growth of a smaller particle at the expense of a larger, should occur. This has been found by Johnson (1984) for the case of two elastically inhomogeneous particles which were constrained to be of a spherical shape. This same restriction has been used in dynamical descriptions of the coarsening process which support the above mentioned finding (Johnson et al., 1990; Johnson and Abinandanan, 1993a, b). However, some investigations claim, that the restriction placed on the particle shapes can lead to qualitatively incorrect answers. Specifically, Su and Voorhees (1996a, b) showed for the elastically homogeneous case that, while two spherical particles can be a minimizer of the functional (2), two particles of unconstrained shape are never stable with respect to coarsening: An – even only slightly – larger particle will always grow at the expense of the smaller. This is consistent with our findings, as in the homogeneous case all solutions have been found to be unstable. But our results show that two separate particles can indeed be an energetic minimum – and should therefore be stable against coarsening – when the elastic inhomogeneity is taken into account. Generally, an increase of the stiffness ratio \( \mu^-/\mu^+ \) as well as a decrease of the inclusions anisotropy \( A^- \) tends to stabilize a two particle system.

The direct calculation of the energies for two particles of unequal size which are otherwise unconstrained revealed an extremely weak dependence of the energy on the ratio of particle sizes. This indicates that growth rates of particles should be very small – especially in the transition region from stable to unstable \((L \approx 12\ldots15)\). These results also suggest that if both particles of a two particle
system were to grow, i.e. $L$ would increase, they might exhibit normal coarsening in the beginning but turn to inverse coarsening when a certain size has been reached.

A particularly interesting result is that, for certain parameter values, there is no repulsive force between two particles, regardless of the interparticle distance (e.g. for $A^+ = 2$, Fig. 12). Here, one should therefore expect a mechanism for the formation of one particle out of two to be operative, that is different from the growth of one particle at the expense of the other: namely the coalescence of the two particles. While it is clear that the kinetics of the process are important for this phenomenon, our results show that elastic inhomogeneity should also be taken into account.

Compared to the other material parameters, the influence of the inclusions average Poisson number $\nu$ is less pronounced. This is due to the fact that, for elongated particles, the deviatoric part of the strain energy prevails inside the inclusion (Thompson et al., 1994; Schmidt, 1997). The deviatoric part of the strain energy can be written in dimensionless form as

$$W_d = (C_{11} - C_{12})(\epsilon_x - \epsilon_y)^2/4 + 2C_{44}\nu^2. \tag{22}$$

Since both $C_{11} - C_{12} = 4/(1 + A)$ and $C_{44} = 2A/(1 + A)$ are independent of $\nu^-$ the results are less dependent on the value of $\nu^-$. The results are even less dependent on the matrix' Poisson number $\nu^+$: within the accuracy of the numerical calculation, no dependence could be noticed. This is because the volumetric strain in the matrix due to the eigenstrain in the inclusion is several orders of magnitude smaller than the deviatoric strain and, hence, the volumetric part of the strain energy can be neglected compared to the deviatoric part. This holds for arbitrary inclusion shapes. Because the volumetric part of the strain energy can be expressed as

$$W_v = (C_{11} + C_{12})\epsilon_v^2/4, \tag{23}$$

$\epsilon_v = \epsilon_1 + \epsilon_2$ being the volumetric strain, field quantities and driving force $\tau_n$ should be independent of $C_{11}^- + C_{12}^- = (3 - 4\nu^+)//(1 - 2\nu^+)$, and hence independent of $\nu^+$.

6. Conclusions

The influence of elastic inhomogeneity on equilibrium and stability of a two particle morphology has been investigated by calculating energy extremizing shapes for a system of two orthotropic materials with an isotropic, constant interfacial energy density. The basic findings of this study are:

- We found three different types of solutions, which correspond to the three solution branches of a disturbed bifurcation diagram for the case of a single particle. In all three solution types the two particles are mirror-symmetry-related.
- The geometry of these shapes is essentially that of a single particle that exhibits an eigenstrain with a small deviatoric part.
- For material parameters close to that of Nickel, most of the two particle morphologies were found to be unstable with respect to changes in their size – implying a tendency of the system to form one large particle out of two by growing one at the expense of the other.
- A different mechanism for the formation of one large particle – particle merging – should be expected for small stiffness ratios, large inclusion anisotropies or small matrix anisotropies, because there is no repulsive force between the particles.
- Both modes of instability are sensitive to the material parameters.
- A two particle system has been proved to be stable by an eigenvalue analysis as well as by direct energy calculation. This situation implies the possibility of inverse coarsening and occurs for small inclusion anisotropies and large stiffness ratios.
- Equilibrium morphologies and their stability have been found to be practically independent of the average matrix' Poisson number.

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Appendix A. Connection of the material parameters with the Voigt-constants

Writing Hookes law for an orthotropic material with cubic symmetry in plane strain as

\[
\begin{bmatrix}
\sigma_x \\
\sigma_y \\
\tau_{xy}
\end{bmatrix} = \begin{bmatrix}
C_{11} & C_{12} & 0 \\
C_{12} & C_{22} & 0 \\
0 & 0 & C_{44}
\end{bmatrix} \begin{bmatrix}
\varepsilon_x \\
\varepsilon_y \\
2\varepsilon_{xy}
\end{bmatrix},
\]

the corresponding values of the \(C_i\) are given by:

\[
C_{11} = C_{22} = \mu \left( \frac{2 \beta + A}{1 + A} - \frac{1 - 4\nu}{1 - 2\nu} \right),
\]

\[
C_{12} = \mu \left( \frac{4\nu}{1 + A} - \frac{1 - 4\nu}{1 - 2\nu} \right),
\]

\[
C_{44} = \mu \frac{2\beta}{1 + A}.
\]

(\(x \) and \(y \) denote the [1 0 0] and [0 1 0] direction of the material respectively).

References


